

A straightforward quantum-mechanical derivation of the Crooks fluctuation theorem and the Jarzynski equality

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We obtain the Crooks and the Jarzynski non-equilibrium fluctuation relations using a direct quantum-mechanical approach for a finite system that is either isolated or coupled not too strongly to a heat bath. These results were hitherto derived mostly in the classical limit. The two main ingredients in the picture are the time-reversal symmetry and the application of the first law to the case where an *agent* performs work on the system. No further assumptions regarding stochastic or Markovian behavior are necessary, neither a master equation or a classical phase-space picture are required. The simplicity and the generality of these non-equilibrium relations are demonstrated, giving very simple insights into the Physics.

I. INTRODUCTION

In contrast to the situation in equilibrium statistical physics, and linear response theory, there are not so many well-established results for systems far from equilibrium [1–3]. Two such extremely interesting results are the “nonequilibrium fluctuation theorem” (NFT) of Crooks [4] and the related Jarzynski equality [5]. Both have to do with the work done by/on a finite system coupled to a heat bath. We also mention here previous works [6–8], showing that the Kubo formalism, the fluctuation-dissipation theorem, and the associated detailed-balance relations are valid in a large class of nonequilibrium steady-state systems, and not only in equilibrium.

The system under consideration is described by a time dependent Hamiltonian $\mathcal{H}(X(t))$, where the parameter X is a time-dependent c-number, often coupled linearly to an observable of the system. At $t = t_0$ the system is prepared in thermal equilibrium at the temperature T . The thermalization is achieved by connecting it for a long enough time to a thermal bath at that temperature. After that, within the time $t = t_1$, the system undergoes a “work process”. This means that an “agent” changes the value of X from X_0 to X_1 . During this process a work \mathcal{W} is performed on the system, and possibly some heat \mathcal{Q} is dissipated into the bath [9]. In the simplest scenario the system is isolated, and heat flow is not involved. It should be emphasized that at the end of the work process, the system is in general *not* in equilibrium.

The NFT deals with the probability distribution $P(\mathcal{W})$ of the work \mathcal{W} , whose experimental determination requires to repeat the process protocol many times, and to record the measured values of \mathcal{W} . Specifically the NFT concerns the ratio $P_{0 \rightarrow 1}(\mathcal{W})/P_{1 \rightarrow 0}(-\mathcal{W})$ between the statistics of the forward scenario, and the statistics of the reversed scenario. In the latter, the system is equilibrated with the same bath under conditions such that $X = X_1$. Then the time-reversed process protocol is realized, such that at the final time $X = X_0$.

The objective of this paper is to present a simple derivation of the NFT in the quantum mechanical context, bypassing various subtleties that, in our view, have

obscured the simple physics involved. The main issue is to define carefully the notion of work in the quantum mechanical context, and to clarify the role that is played by the bath.

Outline: We refer to the evolution during a work process, and formulate for it a generalized detailed balance relation. Then we discuss the notion of work, leading to the NFT of Crooks. The main issue is the modeling the work agent, and the understanding of the role that is played by the bath. The implied Jarzynski equality and the implications on the wasted work and on the entropy production are briefly discussed.

II. EVOLUTION DURING A WORK PROCESS

The system under consideration is described by a time-dependent Hamiltonian $\mathcal{H}(X(t))$. Let us assume that a classical “agent” changes the value of the c-number control parameter X from X_0 at $t = t_0$ to X_1 at $t = t_1$. In some cases, but not in general, the actual duration of the time dependent stage might be $\tau \ll |t_1 - t_0|$. Say that at $t = t_0$ the system was prepared in some eigenstate $n^{(0)}$ of $\mathcal{H}(X_0)$. Then we can ask what is the probability $P_{0 \rightarrow 1}(m|n)$ that at the later time $t = t_1$ it is measured in an eigenstate $m^{(1)}$ of $\mathcal{H}(X_1)$. Below we use the notation

$$\omega = E_m^{(1)} - E_n^{(0)} \quad (1)$$

In a later section we shall define the notion of work \mathcal{W} , and shall explain that up to some uncertainty, we can make the identification $\mathcal{W} = \omega$, provided the system is isolated from the environment.

For a strict quantum adiabatic process one has $P(m|n) = \delta_{n,m}$. But we are interested in more general circumstances. In particular we focus in this section on unitary evolution for which

$$P_{0 \rightarrow 1}(m|n) = \left| \langle m^{(1)} | U_{0 \rightarrow 1} | n^{(0)} \rangle \right|^2, \quad (2)$$

where U is the time-evolution operator. What is important for the derivation of the NFT is the micro-

reversibility of the dynamics, namely,

$$P_{1 \rightsquigarrow 0}(n|m) = P_{0 \rightsquigarrow 1}(m|n) \quad (3)$$

Note that in general the reversed process requires to transform some fields, e.g. to change the sign of the magnetic field if present.

For completeness it is also useful to define the notion of “classical dynamics”. Given phase-space, we can divide it into cells in some arbitrary desired resolution. Then we can regard n as an index that labels cells in phase space. The classical equations define a map

$$|n_{final}^{(1)}\rangle = M |n_{initial}^{(0)}\rangle \quad (4)$$

We use quantum style notations in order to make the relation to the quantum formulation clear. It follows that $P(m|n)$, instead of being a stochastic kernel, becomes a deterministic kernel that induces *permutations*

$$P_{0 \rightsquigarrow 1}(m|n) \Big|_{\text{classical}} = \delta_{m, Mn} \quad (5)$$

The derivation in the next section does not depend on whether the dynamics is “classical” or “stochastic” or “quantum” in nature as long as the *measure* and the micro-reversibility are preserved. The preservation of *measure* is reflected by our discrete notations: If, say, we had deterministic dynamics that does not satisfy Liouville theorem, we could not have used the above “cell construction”

III. THE GENERALIZED DETAILED BALANCE RELATION

The power spectrum of the fluctuations of an observable A is given by the following spectral decomposition

$$\tilde{S}(\omega) = \sum_{n,m} p_n \left| \langle m|A|n \rangle \right|^2 \delta(\omega - (E_m - E_n)) \quad (6)$$

Here we assume a time independent Hamiltonian, and stationary preparation that can be regarded as a mixture of eigenstates with weights p_n . For a canonical preparation

$$p_n = \frac{1}{Z} e^{-E_n/T} = \exp \left[-\frac{E_n - F(X_0)}{T} \right] \quad (7)$$

where Z is the partition function, and $F(X)$ is the Helmholtz free energy at temperature T , calculated here for the fixed value of the control parameter X . Then one obtains after two lines of straightforward algebra, the detailed balance relation

$$\frac{\tilde{S}(\omega)}{\tilde{S}(-\omega)} = \exp \left[\frac{\omega}{T} \right] \quad (8)$$

This relation plays a key role in the linear response theory. Specifically it reflects the ratio between the tendency

of the system to absorb and emit energy from/to a driving source $-f(t)A$.

In complete analogy we define the following spectral kernel:

$$\begin{aligned} P_{0 \rightsquigarrow 1}(\omega) &= \sum_{n,m} p_n^{(0)} P_{0 \rightsquigarrow 1}(m|n) \delta(\omega - (E_m^{(1)} - E_n^{(0)})) \end{aligned} \quad (9)$$

Here the superscript indicates whether we refer to the initial Hamiltonian $\mathcal{H}(X_0)$ or to the final Hamiltonian $\mathcal{H}(X_1)$. For the reversed process we write

$$\begin{aligned} P_{1 \rightsquigarrow 0}(\omega) &= \sum_{m,n} p_m^{(1)} P_{1 \rightsquigarrow 0}(n|m) \delta(\omega - (E_n^{(1)} - E_m^{(0)})) \end{aligned} \quad (10)$$

It immediately follows, in analogy with the usual detailed balance condition, that the ratio of the spectral functions $P_{0 \rightsquigarrow 1}(\omega)$ and $P_{1 \rightsquigarrow 0}(-\omega)$ is determined by the ratio of the initial probabilities $p_n^{(0)}$ and $p_n^{(1)}$, leading to

$$\frac{P_{0 \rightsquigarrow 1}(\omega)}{P_{1 \rightsquigarrow 0}(-\omega)} = \exp \left[\frac{\omega - (F(X_1) - F(X_0))}{T} \right], \quad (11)$$

where both $F(X_1)$ and $F(X_0)$ refer to the same preparation temperature T . Note again that if X does not change in time, this relation formally coincides with the detailed balance relation Eq.(8).

IV. THE NOTION OF WORK AND THE NFT OF CROOKS

The main difficulty in the quantum formulation of the NFT concerns the definition of work. Consider first an isolated system. Naively we can define $\mathcal{W} = \omega$, namely the work is the change in the energy of the system. But in the quantum reality this means that we have to do a measurement of the initial stage, hence the state of the system collapses and it is no longer canonical.

Furthermore, assume that we want to consider a multi-stage process that extends over two time intervals $t_0 \rightarrow t_1 \rightarrow t_2$. We would like to say that the work done is the sum $\mathcal{W}_{0 \rightsquigarrow 1} + \mathcal{W}_{1 \rightsquigarrow 2}$. With the above definition we have to perform a measurement at the time instant t_1 . But in the quantum mechanical reality we do not have the time for that...

It is therefore clear that the definition of work requires refinement. One possible direction [10] is to define

$$\mathcal{W}_{0 \rightsquigarrow 1} = \int_{t_0}^{t_1} \frac{\partial \mathcal{H}}{\partial X} \dot{X} dt \quad (12)$$

Then, in analogy with the theory of *counting statistics* [11–13], one might say that a *continuous measurement* is required, involving a weak coupling to a von Neumann pointer. The problem with this approach is that the

counting statistics *quasi probability* [12, 13] has no simple physical interpretation, and might be even negative.

It turns out that in the present context there is a simple way out of these subtleties. Instead of regarding $X(t)$ as a c-number field, we regard it as a dynamical variable of an “agent” that is doing work. The total Hamiltonian can be formally written as

$$\mathcal{H}_{\text{total}} = \mathcal{H}(r; X) + \mathcal{H}_{\text{agent}}(X) \quad (13)$$

where r stands for system dynamical variables, and X is the agent degree of freedom. Then we define the work \mathcal{W} as the change in the energy of the agent

$$\mathcal{W} = E_{\text{agent}}(0) - \mathcal{H}_{\text{agent}} \quad (14)$$

where $E_{\text{agent}}(0)$ is its initial energy, which is assumed to be well defined up to some small uncertainty. In the next section we clarify that for an isolated system $\mathcal{W} = \omega$, to the extent that the above small uncertainty can be ignored, as should be the case for a classical agent. From here follows the Crooks relation

$$\frac{P_{0 \rightarrow 1}(\mathcal{W})}{P_{1 \rightarrow 0}(-\mathcal{W})} = \exp \left[\frac{\mathcal{W} - (F(X_1) - F(X_0))}{T} \right] \quad (15)$$

Instead of going on with an abstract discussion of what do we mean by “work agent”, we consider in the next section two simple prototype models that illuminate this notion.

V. MODELING THE WORK AGENT

In order to define the notion of work we find it essential to regard the “agent” as a dynamical entity. It can be another object (“piston”) from/to which energy is transferred, or it can be a field with which the system interacts, absorbing or emitting excitations (“photons”).

A. Modeling the agent as a piston

The prototype model for explaining the notion of work in standard thermodynamics textbooks is the gas-piston system that is illustrated in Fig.1. The “agent” on which work is being done is a piston that is free to move to infinity. After the piston is pushed out the gas particles stay in the box, and no longer interact with the piston, but possibly may interact (say) with a bath or with other agents. At the end of each single “run” of the experiment, there is an unlimited time to measure the energy of the freely moving piston in the desired resolution.

The essential ingredient in the illustrated construction is the decoupling at the end of the interaction: After the piston moves outside of the shaded region, it becomes a free object whose kinetic energy we can measure without having a time limitation.

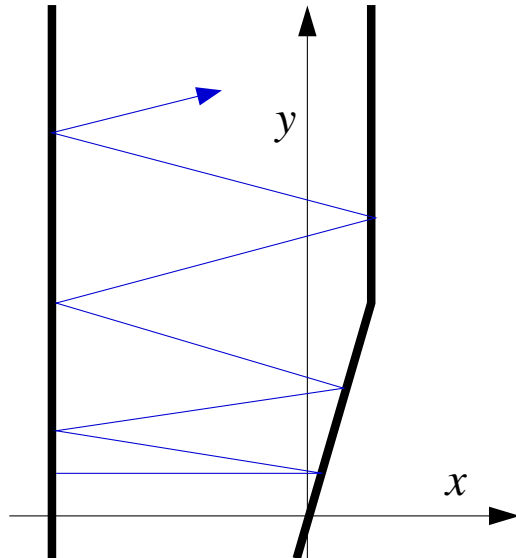
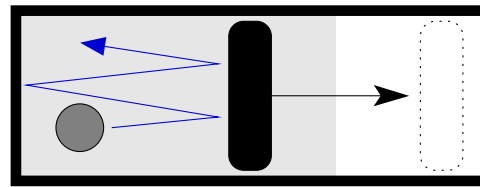


FIG. 1: The system is a gas of particles in a box. The box region is indicated by grey. A representative trajectory is illustrated. The “agent” on which work is being done is a piston that is free to move to infinity. After the piston is pushed out the gas particles stay in the box, and no longer interact with the piston, but possibly may interact (say) with a bath or with other agents. At the end of each single “run” of the experiment, there is an unlimited time to measure the energy of the freely moving piston in the desired resolution.

For presentation purposes, but without any loss of generality, we consider a single gas particle and regard the box as one dimensional. The Hamiltonian is

$$\mathcal{H}_{\text{total}}(r, p; X, P) = \frac{p^2}{2m} + V_{\text{box}}(r) + u(r - X) + \frac{P^2}{2M} \quad (16)$$

where $u(r - X) = u_0 \delta(r - X)$ with $u_0 = \infty$. Thanks to a potential V_{box} the gas particle remains in the shaded region even if the piston is “out”. Once the piston is out the “system” no longer affects the “agent”, nor affected by it.

In order to visualize the dynamics it is convenient to define $\alpha = (m/M)^{1/2}$, and $p_x = p$, and $p_y = \alpha P$, and $x = r$, and $y = (1/\alpha)X$. Then the Hamiltonian takes the form

$$\mathcal{H}_{\text{total}} = \frac{1}{2m}(p_x^2 + p_y^2) + V_{\text{box}}(x) + u(x - \alpha y) \quad (17)$$

We assume that initially the piston is prepared in rest with some uncertainty ΔX in its position, and an asso-

ciated uncertainty $\Delta P \sim 1/\Delta X$ in its momentum. Accordingly the uncertainty of the total energy is

$$\Delta E \sim [M\Delta X^2]^{-1} + \Delta E_{\text{system}} \quad (18)$$

The total Hamiltonian is a constant of motion. It follows that the probability distribution of the total energy E is a δ -function. The total energy is the sum of the particle energy and the piston energy. Let us denote the increase in the particle energy as ω , and the decrease in the piston energy as \mathcal{W} . It follows that the joint distribution is

$$P(\omega, \mathcal{W}) = P(\omega) \delta(\mathcal{W} - \omega) \quad (19)$$

where the equality is justified to the extent that ΔE can be neglected. Under such conditions the distribution of work $P(\mathcal{W})$ is the same as $P(\omega)$.

The argument above has established the equality of $P(\mathcal{W})$ and $P(\omega)$ for a system that is prepared in a microcanonical state, such that ΔE_{system} is a small uncertainty. But trivially the equality of the two distributions extends to any mixture, and in particular to the canonical preparation under consideration. We note that our definition of $P(\omega)$ in the previous section has assumed a c-number driving source, while here there is some uncertainty ΔX in the position of the piston. Accordingly a trade-off is required with regard to ΔX and δE . This trade-off is the physical limit of the NFT applicability. In practice, and in particular for large deviations, this uncertainty should not be an issue.

B. Modeling the agent as a field

In this subsection we consider another illuminating example for a “work agent” but with a different emphasis: here the agent is a field with which the system interacts, and the measurement is the detection of a field quanta. The reasoning below is based initially on the traditional weak coupling assumption.

Consider a classical force \mathcal{F} that arises, say, from a classical electric field that acts on charged particles. Taking the coupling to be via the total dipole moment of the system, the interaction term is

$$\mathcal{H}_{\text{system-agent}} = -\mathcal{F}(t) \sum r_i \quad (20)$$

where the r_i are the coordinates of the particles along the relevant axis, and $\mathcal{F}(t)$ is a c-number force that is switched from $\mathcal{F}_0 = 0$ at $t_0 = 0$, to $\mathcal{F}_1 = \delta\mathcal{F}$ at time t_1 .

To see what is going on, think of expanding $\mathcal{F}(t)$ in a Fourier integral. the Fourier components \mathcal{F}_ω are significant on an interval of order $1/\tau$, where τ is the actual duration of the variation, which is possibly small compared with $|t_1 - t_0|$. Small $\delta\mathcal{F}$ and/or small τ make the relevant Fourier components small. From low order perturbation theory it follows that the transitions are to levels E_m whose energy is within $\sim 1/\tau$ of the initial energy E_n , with probabilities proportional to $|\mathcal{F}_\omega|^2$. Very

importantly, energy is conserved in the sense that the excitation takes an energy $\omega = E_m - E_n$ from the field [14].

While we employed here perturbation-theory thinking, these considerations are much more general. For stronger perturbations, one may think in terms of the evolution operator, propagating the system in time. In particular, the symmetries [15] due to microscopic reversibility, will follow *mutatis mutandis*.

One should not worry about the time-energy uncertainty even in the short τ limit, the relevant scale for the NFT considerations is t_1 that can be much larger. We know that if we quantize the field \mathcal{F} , a photon with the energy $\omega = E_m - E_n$ will be destroyed during the transition.

In the absence of a coupling to the bath, let us assume that τ is small such that its inverse is larger than the mean level spacing. Under such circumstances the transitions are into an energy range that contains many states, within an interval whose spectral width is $1/\tau$ for short pulse, that saturates to a limiting value Γ_F for long pulse. In the regime where first order-perturbation theory applies Γ_F becomes much smaller compared with the mean level spacing of the system. When a coupling to the bath is introduced, the levels of the system acquire an additional width Γ_B . If the interaction is weak enough Γ_B would also become smaller than the mean level spacing of the *system*. Under such circumstances the sharp levels become narrow well-separated peaks in the system density of states.

For the reverse process, the lineshape around the levels in the final stage have in general not the same width as that of the corresponding peak in the forward process, but, by T symmetry, its total weight is the same, and this is what counts in obtaining Eq.(11). Energy conservation then implies that $\mathcal{W} = \omega$, so that again the distributions of ω and of \mathcal{W} are the same. This again establishes Eq.(15).

VI. THE IRRELEVANCE OF THE BATH

The Crooks relation and the Jarzynski equality concern the probability distribution of *work* done during non-equilibrium process that starts with a canonical state. We deduced in the previous sections that in the case of an isolated system $P(\mathcal{W})$ satisfies the same Crooks relation as $P(\omega)$. We now want to extend the validity of this relation to the case of non-isolated system. Inspired by the approach of [5], let us discuss whether an interaction with a bath can affect these relations.

First scenario.— After the work process has ended we allow the system to relax to the bath temperature T . This additional step does not involve work, hence $P(\mathcal{W})$ is not affected.

Second scenario.— Assume that there is a finite system-bath coupling η during the process. The duration of the process is τ . Regarding the system and the

bath as one grand-system we write

$$\frac{P_{0 \rightsquigarrow 1}(\mathcal{W}; \eta, \tau)}{P_{1 \rightsquigarrow 0}(-\mathcal{W}; \eta, \tau)} = \exp \left[\frac{\mathcal{W} - (F_{tot}(X_1; \eta) - F_{tot}(X_0; \eta))}{T} \right] \quad (21)$$

It should be clear that $P_{0 \rightsquigarrow 1}(\mathcal{W})$ and $P_{1 \rightsquigarrow 0}(-\mathcal{W})$ depend on both η and τ . But the ratio, according to Crooks, is independent of τ . Still one suspects that the right-hand side depends on η . But in fact this is not so. The argument is as follows: The ratio is independent of τ , and therefore we can evaluate it, without loss of generality, for $\tau \rightarrow 0$; But in this "sudden" limit the result should be independent of η , because the bath has no time to influence the work. We therefore can set $\eta = 0$, and deduce that without loss of generality

$$\frac{P_{0 \rightsquigarrow 1}(\mathcal{W}; \eta, \tau)}{P_{1 \rightsquigarrow 0}(-\mathcal{W}; \eta, \tau)} = \exp \left[\frac{\mathcal{W} - (F_{sys}(X_1) - F_{sys}(X_0))}{T} \right] \quad (22)$$

without dependence on η and τ . Hence the NFT is established for a process in which the system is non-isolated. In particular, it may interact with a thermal reservoir.

VII. THE JARZYNSKI EQUALITY

It is well known [4] that the Jarzynski equality [5] is an immediate consequence that follows from the Crooks relation Eq.(15). For completeness we repeat this derivation here. Multiplying both sides of the Crooks relation by $P_{1 \rightsquigarrow 0}(-\mathcal{W})$, integrating over \mathcal{W} , and taking into account the normalization of $P(\mathcal{W})$, one obtains

$$\left\langle \exp \left[-\frac{\mathcal{W}}{T} \right] \right\rangle = \exp \left[-\frac{F(X_1) - F(X_0)}{T} \right], \quad (23)$$

which is the Jarzynski relation. From here follows that

$$\langle \mathcal{W} \rangle > F(X_1) - F(X_0) \quad (24)$$

This variation of the 2nd law of thermodynamics is known as the *maximum work principle*, because it sets an upper bound on the work $W = -\mathcal{W}$ that can be *extracted* from a work process. Optionally it can be regarded as the minimum work \mathcal{W} needed from the agent to do the process [9]. Note that our sign conventions for \mathcal{W} and \mathcal{Q} are opposite to those that are used in most textbooks.

VIII. WASTED WORK AND ENTROPY PRODUCTION

It is instructive to recast the Crooks relation Eq.(15) in terms of entropy produced, as in fact was originally formulated by Crooks. From Eq.(24) it follows that the

difference $\Delta F = F(X_1) - F(X_0)$ is the minimum work that is required in a reversible quasi-static process. Accordingly the difference $\mathcal{W} - \Delta F$ can be regarded as the wasted work in a realistic process. Dividing by T we get a quantity ΔS_w that we can regard as the wasted entropy (For the temperature we use units such that the Boltzmann constant is unity). Consequently the fluctuation theorem Eq.(15) reads:

$$\frac{P_{0 \rightsquigarrow 1}(\Delta S_w)}{P_{1 \rightsquigarrow 0}(-\Delta S_w)} = \exp [\Delta S_w] \quad (25)$$

Below we would like to better clarify the connection with thermodynamics, and in particular with the Clausius version of the 2nd law.

Taking a puristic point of view, one defines thermodynamic functions only for equilibrium states. Therefore let us assume that the system ends up in a thermodynamic equilibrium, say by allowing it to relax at the end of the driving process. Under this assumption we can associate with the initial and final states well defined values of system entropy, whose difference can be expressed using thermodynamic functions:

$$\Delta S = \frac{\Delta E - \Delta F}{T} \quad (26)$$

where by the first law of thermodynamics the change in the energy of the system is

$$\Delta E = \mathcal{W} - \mathcal{Q} \quad (27)$$

The total entropy change of the universe is the sum of the system entropy change, and that of the bath

$$\mathcal{S} = \Delta S + \frac{\mathcal{Q}}{T} = \frac{\mathcal{W} - \Delta F}{T} \quad (28)$$

It follows that the Crooks relation can be written as

$$\frac{P_{0 \rightsquigarrow 1}(\mathcal{S})}{P_{1 \rightsquigarrow 0}(-\mathcal{S})} = \exp [\mathcal{S}] \quad (29)$$

As in the case of the Jarzynski equality we deduce that

$$\left\langle \exp [-\mathcal{S}] \right\rangle = 1 \quad (30)$$

and consequently

$$\langle \mathcal{S} \rangle > 0, \quad (31)$$

in accordance with the second law of thermodynamics. Note that it is only the average $\langle \mathcal{S} \rangle$ that is positive. In a finite system \mathcal{S} is negative for a fraction of the processes, with vanishing manifestation in the thermodynamic limit.

IX. SUMMARY

The objective of this work was to illuminate that the simplicity of the NFT is maintained also in the quantum context. The way to go was to regard it as arising from a generalized detailed balance relation Eq.(11). A key issue was to regard the work agent as a dynamical entity, and to avoid a continuous measurement scheme for its measurement. It was also important to clarify the role of the environment, and to show that neither a master equation nor a classical phase-space picture are required in the derivation. It is possible, and quite instructive, to repeat all considerations in terms of the combined states of the system and bath, in the manner suggested for ex-

ample by Fano [16] and Lipkin [17].

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 - [14] For a closed system, the work done by the classical agent

is all converted to a change of the system's energy. A well-known even stronger example is that of a probe particle inelastically scattered from the system losing an certain energy which is then equal to the energy of the created excitation(s).

- [15] This symmetry follows noting that $W_{ij} = |U(t)_{ij}|^2$ and $W_{ji} = |U(t)_{ji}^{-1}|^2 = |U(t)_{ij}|^2 = W_{ij}$, where $U(t)$ is the (unitary) operator that propagates the system by the time t .
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